

The present invention relates to a process for oligomerizing C_n -olefins, in particular for preparing C_{12} -olefins by dimerization.

As described in the specification beginning at page 1, line 8, processes for the oligomerization of olefins are known, such as that disclosed in DE-A-4339713, which is the German equivalent of Vicari et al. As described in the specification herein at page 1, lines 9-14, Vicari et al. describes a process for oligomerizing olefins to give highly linear oligomers. In this process, C_{2-6} -olefins are reacted at superatmospheric pressure and elevated temperature over a fixed-bed catalyst comprising from 10 to 70% by weight of nickel oxide, from 5 to 30% by weight of titanium dioxide and/or zirconium dioxide, from 0 to 20% by weight of aluminum oxide as significant active constituents and silicon dioxide as the remainder.

As described in the specification at page 1, line 25, a disadvantage of the known processes is that the catalyst life is frequently too short. The catalyst is, in particular, clogged by higher oligomers and therefore loses its activity.

Applicants have discovered that by oligomerizing C_n -olefins using the above-discussed fixed-bed catalyst of Vicari et al. at a conversion to oligomerized C_n -olefins of not more than 30% by weight (now from 10 to 30% by weight), based on the reaction mixture, deactivation of the catalyst can be avoided and dimer selectivity can be increased. By recycling unreacted reaction mixture after separating off the oligomers, a higher total conversion can be obtained. Indeed, the presently-claimed process makes it possible to realize a total conversion of over 90% together with a C_{12} (dimer) selectivity of over 80%.

Adherence to the conversion specified according to the claims greatly increases the operating life of the catalyst, since the formation of high-boiling compounds which can deposit on the catalyst and thus cause a drop in activity is suppressed, as described in the specification at page 2, lines 22-27.

During the a previous interview, the Examiner commented that there is no data in the specification demonstrating increased catalyst operating life. Applicants' reply was that since the Examiner has provided no evidence to suggest that catalyst operating life is not improved by the present invention, Applicants' have no burden to disprove such evidence. Moreover, Applicants, their attorneys, and all associated with the filing and prosecution of the application are under a duty of candor and good faith under 37 CFR 1.56. The Examiner has no reason to question the truth of Applicant's assertion regarding catalyst life.

While Vicari et al discloses the presently-recited fixed-bed catalyst, as discussed above, Vicari et al do not recognize the nexus between maximum conversion, on the one hand, and increased catalyst life, higher selectivity of dimer, and higher overall conversion, --- on the other hand.

Examples 1 and 2 illustrate the presently-claimed invention, and the above-discussed superior results. In Example 1, beginning with a hydrocarbon mixture feed containing 73% by weight of C_6 component, a C_6 -olefin conversion of 94.7% and a C_{12} selectivity of 83.6%, based on the C_6 -olefins reacted, was obtained.

In Example 2, the C_6 concentration in the hydrocarbon mixture feed was 98.7% by weight. The result was a C_6 -olefin conversion of 98.4% and a C_{12} selectivity of 82.6%, based on the C_6 -olefins reacted.

During the above-referenced previous interview, the Examiner queried how the results in Examples 1 and 2 square with the above-discussed limitation of a maximum conversion of not more than 30% by weight. Applicants' attorney and the assignee's representative explained, referring to Figs. 1A or 1B, that the 30% conversion is with respect to the input to, and output from, reactor C1, while the final conversion and selectivity of the Examples is with respect to bottoms stream S. The conversion regarding input to and output from C1 for

each Example was calculated and is 20.2% for Example 1, and 19.4% for Example 2, as disclosed in the specification in the "*Total C₄*," column and "Feed mixture to K1=reactor output" row, at the top of page 9, and the bottom of page 10, respectively.

As discussed above, Vicari et al do not recognize any nexus between conversion and the other listed properties. Indeed, based on the examples and comparative examples in Vicari et al, the conversion varies from 41-55% for the comparative examples, and 53-78% for the examples. See the various tables at column 7 therein. Since no recycle is disclosed, it is assumed that these conversions are ultimate conversions. While this data is for C₄-olefins, nevertheless, it is still sufficient to show that Vicari et al do not recognize the significance of maintaining the conversion below a certain maximum for each single pass over the catalyst.

Sanderson et al does not remedy any of the above-discussed deficiencies of Vicari et al. Sanderson et al discloses a process for oligomerizing olefins using sulfate-activated molecular sieves. Sanderson disclose that by the use of such molecular sieves, a high percentage of dimers, i.e., a high dimer to trimer ratio, can be achieved (column 2, lines 35-37). Sanderson et al further disclose that the oligomerization results in an oligomer product distribution that varies with reaction time: as the reaction time increases, the olefin monomer conversion increases, and the selectivities for the heavier oligomers increase (column 3, lines 33-55). The olefin monomer feed stocks of Sanderson et al must contain at least 10 carbon atoms (column 3, lines 13-21).

In Sanderson et al, no particular olefin conversion goal is disclosed, although it is noted that liquid hourly space velocity may be varied within wide limits (column 5, lines 55-61), and it is noted that the olefin conversion, as specified in the table bridging columns 7 and 8, varies widely, from 3.0% to 70.4%. Moreover, the data therein show that conversion is not strictly a function of reaction time, as can be seen by comparing examples using the same

catalyst. For example, Catalyst 4 produces a conversion of 3% in 5 hours (Example 1), and a conversion of 11.3% in 4 hours, but at a higher temperature (Example 7). A comparison of Examples 2 and 6; Examples 4 and 5; produces a similar result. Thus, the conversion was actually lower after a longer reaction time. In addition, all the examples therein employed a reaction time of 4 or 5 hours, yet no trend can be discerned therefrom that conversion must be limited to 30% or lower to maximize dimer product. Rather, the trend is due to the use of sulfate-activated molecular sieves (Catalysts 2-4 and 7) compared to the non-sulfate-activated molecular sieves (Catalysts 1, 5 and 6). Compare Examples 3 and 5, where at identical feeds, reaction times and temperatures, the sulfated molecular sieve produced a conversion of 32.5% (Catalyst 2), while the same catalyst in non-sulfated form produced a conversion of 18.8% (Catalyst 1). Compare also Examples 11 and 13, which also shows increased conversion using the sulfated molecular sieve.

Sanderson et al adds nothing to Vicari et al. Sanderson et al is concerned with a different catalyst therefrom, and olefins having a carbon range outside the range specified by Vicari et al. Sanderson et al disclose nothing with regard to any possibility of increased catalyst life by limiting conversion percentage to a particular amount. Nor would one skilled in the art even combine Vicari et al and Sanderson et al in view of the mutually exclusive catalysts and olefins used. Indeed, without the present disclosure as a guide, one skilled in the art would not be motivated to combine Vicari et al and Sanderson et al. Even if combined, the result would not be the presently-claimed invention.

During the above-referenced previous interview, the Examiner took the position, erroneously in Applicants' view, that a demonstration of increased catalyst life was necessary to establish patentability over the applied combination of prior art. While Applicants submitted that they had shown patentability in the absence of such demonstration, for reasons

discussed above, nevertheless, the Heidemann Declaration of record provides such a demonstration.

In the Office Action, the Examiner first responds to the Heidemann Declaration of record, finding, in effect, that not enough information is provided for the processes of Examples 1 and 3. In reply, the Examiner's findings are not understood, since the Experimental Report, attached to the Heidemann Declaration, describes all the relevant parameters.¹

The catalyst used in the Experimental Report was the same catalyst employed in Examples 1 and 2 of the specification herein, as disclosed at page 7, lines 11-14. This catalyst was employed in each of Examples 1-3 of the Experimental Report.² In the Experimental Report examples, Example 1, the reaction temperature was 80°C; in Example 2, the reaction temperature was 160°C; and in Example 3, the reaction temperature was 60°C. The reaction pressure was 15 bars in all examples. Example 1 of the Experimental Report shows that conversion, based on a single path, was around 17%, even after 69 hours. In Example 3, it was around 20%, even after 168 hours. In Example 2, on the other hand, which was for purposes of comparison, the conversion steadily decreased, indicating catalyst deactivation, or stated in other words, a short catalyst life. The conversions of around 17% and around 20% are sufficient to demonstrate patentability of the presently-recited 10 to 30%

¹For the Examiner's convenience, **submitted herewith** is another copy of the Experimental Report. If the Experimental Report was not included as part of the Heidemann Declaration as filed, this was inadvertent. Applicants' counsel represents that the Experimental Report was attached to the Heidemann Declaration when received by counsel.

²The Examiner should note that Examples 1-3 of the Experimental Report are different from Examples 1 and 2 described in the specification, although the same catalyst is used in each.

by weight conversion, even if a *prima facie* case of obviousness had been made out. No such case has been made out, however.

Nor is the Examiner correct that Applicants must compare their process for all possible conversions within the claims, i.e., from above 0 to 30% by weight (now 10 to 30% by weight), to processes wherein the conversion ranges from 30% up to 100%. Since the prior art neither discloses nor suggests any nexus between conversion and other properties, such as catalyst life, Applicants were under no burden to provide **any** comparative data.

The Examiner criticizes the data as not being commensurate in scope with the claims. In reply, data of patentability must be commensurate in scope with only that part of claimed subject matter that is *prima facie* obvious. However, as discussed above, since the prior art does not recognize any nexus between conversion and catalyst life, there is **no** *prima facie* case of obviousness. Moreover, even if there was such a case, the Examiner may not consider such evidence only for its "knockdown ability". As stated in *In re Carleton*, 599 F.2d 1021, 1024, 202 USPQ 165, 168-69 (CCPA 1979) (**copy enclosed**):

If the applicant presents rebuttal evidence, the decision-maker must consider all of the evidence of record (both that supporting and that rebutting the *prima facie* case) in determining whether the subject matter as a whole would have been obvious. [footnote omitted] *In re Rinehart*, 531 F.2d 1048, 189 USPQ 143 (CCPA 1976); *In re Lewis*, 443 F.2d 389, 170 USPQ 84 (CCPA 1971). The correct procedure for considering rebuttal evidence was set forth by this court in *In re Rinehart*, *supra* at 1052, 189 USPQ at 147:

Though the burden of going forward to rebut the *prima facie* case remains with the applicant, the question of whether that burden has been successfully carried requires that the entire path to decision be retraced. An earlier decision should not, as it was here, be considered as set in concrete, and applicant's rebuttal evidence then be evaluated only on its knockdown ability. Analytical fixation on an earlier decision can tend to provide that decision with an undeservedly broadened umbrella effect. *Prima facie* obviousness is a legal conclusion, not a fact. Facts established by rebuttal evidence must be evaluated along with the facts on which the earlier conclusion was reached, not against the conclusion itself. Though the tribunal must

begin anew, a final finding of obviousness may of course be reached, but such finding will rest upon evaluation of all facts in evidence, uninfluenced by any earlier conclusion reached by an earlier board upon a different record.

At page 6, the Examiner finds Applicants' argument that by recycling unreacted mixture after separating off the oligomers, a higher total conversion can be obtained, is not persuasive. The Examiner's reason is that Applicants do not claim this feature. In reply, the Examiner is incorrect. This feature is claimed in Claim 8.

The Examiner has, in effect, dismissed all of the above arguments with regard to Sanderson et al by simply focusing on the disclosure at column 3, lines 33-55 thereof, concluding therefrom that one skilled in the art would vary conversion to optimize selectivity of oligomers. In reply, it is respectfully submitted that the Examiner has grossly simplified the disclosure in Sanderson et al. At any rate, as discussed above, Sanderson et al discloses no nexus between conversion in a single pass and selectivity, and clearly discloses and suggests no nexus between such conversion and catalyst life.

For all the above reasons, it is respectfully requested that the rejection over Vicari et al in view of Sanderson et al be withdrawn.

The rejection of Claim 9 under 35 U.S.C. § 103(a) as unpatentable over Vicari et al in view of Sanderson et al, and further in view of the English Abstract of EP 395857A (Huls), is respectfully traversed. The Examiner relies on Huls for its disclosure of removing catalyst-poisoning components from a hydrocarbon stream. But, even if Huls were combined with Vicari et al and Sanderson et al, it would still not remedy the above-discussed deficiencies in the combination of Vicari et al and Sanderson et al. Accordingly, it is respectfully requested that this rejection be withdrawn.

All of the presently pending claims in this application are now believed to be in immediate condition for allowance. Accordingly, the Examiner is respectfully requested to pass this application to issue.

Respectfully submitted,

OBLON, SPIVAK, McCLELLAND,
MAIER & NEUSTADT, P.C.



Norman F. Oblon
Attorney of Record
Registration No. 24,618

Harris A. Pitlick
Registration No. 38,779



22850

(703) 413-3000
Fax #: (703) 413-2220
NFO:HAP:cja
E:\atty\HAP 213127US-af.wpd

**Marked-Up Copy**

Serial No: 09/914,532

Amendment Filed on
Herewith:IN THE CLAIMS

--1. (Amended) A process for oligomerizing C_6 -olefins by reaction of a C_6 -olefin-containing reaction mixture over a nickel-containing fixed-bed catalyst, comprising from 10 to 70% by weight of nickel oxide, from 5 to 30% by weight of titanium dioxide and/or zirconium dioxide and from 0 to 20% by weight of aluminum oxide as significant active constituents and silicon dioxide as the remainder, wherein the reaction over the fixed-bed catalyst is carried out continuously in the liquid phase and run a conversion to oligomerized C_6 -olefins of [not more than] from 10 to 30% by weight based on a throughput of the reaction mixture through the catalyst in a single pass.

2. (Amended) A process as claimed in claim 1, wherein the reaction over the fixed-bed catalyst is run at a conversion to oligomerized C_6 -olefins from 10 to [30] 25% by weight, based on the reaction mixture.

5. (Cancelled).

6. (Amended) A process as claimed in claim [5] 1 which is carried out adiabatically in a shaft oven and in which part of the reacted mixture is returned to the reaction.--

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Experimental Report

In the following experiments the hex²ene feed and the catalyst employed were the same as defined in example 2 of the present application (US 09/914,532) see page 9 of the specification. The same catalyst was employed as in the examples of the patent application, compare page 7, lines 11 to 14 of US 09/914,532.

Example 1 (according to the present invention):

5 ml of the catalyst (material which had been produced in the form of 5 mm x 5 mm pellets as described in DE-A-43 439 713; composition in % by weight of the active components: 50 % by weight of NiO, 13 % by weight of TiO₂, 34 % by weight of SiO₂, 3 % by weight of Al₂O₃) were placed in a tube reactor having a length of 15 cm and a diameter of 1 cm. A hexene feed purified over an absorber was fed to the catalyst (1 g feed/1 ml of catalyst x h) at a reaction temperature of 80°C (reaction pressure 15 bar). After 30 hrs, 45 hrs and 69 hrs of running time a probe was taken and the conversion was determined: 17.2 % (30 h), 16.9 % (45 h), 16.2 % (69 h).

Example 2 (Comparison)

The same process as in example 1 was carried out, however, the reaction temperature was 160°C. After 22 hrs, 46 hrs and 63 hrs of running time a probe was taken and the conversion was measured: 44.5 % (22 h), 34.7 % (46 h), 28.8 % (63 h).

Example 3 (according to the present invention)

2500 ml of the catalyst as employed in example 1 was placed in five tube reactors having a length of 100 cm and a diameter of 16 mm arranged in a series. Hexene feed which had been prepurified over an absorber was introduced in the catalyst (0.05 g feed/1 ml catalyst x h) at a reaction temperature of 60°C (operating pressure 15 bar). After 72 hrs, 120 hrs and 168 hrs of running time probes were taken and the conversion was determined: 20.0 % (72 h), 20.2 % (120 h), 19.8 % (168 h).

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In re Carleton, 202 USPQ 165 (CCPA 1979)

In re Carleton

**(CCPA)
202 USPQ 165**

Decided June 7, 1979

No. 78-634

U.S. Court of Customs and Patent Appeals

Headnotes

PATENTS

1. Board of appeals -- Issues determined (§ 19.30)

Patentability -- Invention -- In general (§ 51.501)

Patentability -- Invention -- Law or fact question (§ 51.507)

Pleading and practice in courts -- Burden of proof -- In general (§ 53.131)

Pleading and practice in Patent Office -- Rejections (§ 54.7)

Prior adjudication -- Applications for patent (§ 56.05)

Burden of proof is on Patent and Trademark Office to establish prima facie

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case of obviousness for rejection under 35 U.S.C. 103, and burden of going forward with evidence to rebut that prima facie case then shifts to applicant; whether prima facie case is "strong" or "weak" is not material; once applicant presents rebuttal evidence, decision maker must consider all evidence of record in determining whether subject matter as a whole would have been obvious; question of whether applicant's burden has been successfully carried requires

that entire path to decision be retraced; earlier decision should not be considered as "set in concrete," and applicant's rebuttal evidence then evaluated on its "knockdown" ability; analytical fixation on earlier decision can tend to provide that decision with undeservedly broadened effect; obviousness is legal conclusion based on factual evidence, and not factual determination; facts established by rebuttal evidence must be evaluated along with facts on which earlier conclusion was reached, not against conclusion itself; proper issue on appeal from rejection is whether board erred, as matter of law, in holding that claims were properly rejected; in deciding this issue, court will make independent determination as to legal conclusions and inferences that should be drawn from findings of fact; although tribunal must begin anew, final finding of obviousness will rest upon all facts in evidence, uninfluenced by earlier conclusion reached by earlier board upon different record.

2. Patentability -- Composition of matter (§ 51.30)

Patentability -- Invention -- Specific cases -- Chemical (§ 51.5093)

Although there is vast amount of knowledge about general relationships in chemical arts, chemistry is still largely empirical, and there is often great difficulty in predicting precisely how a given compound will behave; while analogy is at times useful, both organic and inorganic chemistry are essentially experimental sciences, and results are often uncertain, unpredictable, and unexpected.

Particular patents -- Hydroquinone Production

Carleton, Process, rejection of claims 9-13 and 15-16 reversed.

Case History and Disposition:

Appeal from Patent and Trademark Office Board of Appeals.

Application for patent of Peter S. Carleton, Serial No. 563,464, filed March 31, 1975. From decision rejecting claims 9-13 and 15-16, applicant appeals. Reversed.

Attorneys:

Roman Saliwanchik, Kalamazoo, Mich. (Denis A. Firth, North Haven, Conn., and John Kekich, Kalamazoo, Mich., of counsel) for appellant.

Joseph F. Nakamura (Fred W. Sherling, of counsel) for Commissioner of Patents and

Trademarks.

Judge:

Before Markey, Chief Judge, Rich, Baldwin, and Miller, Associate Judges, and Kunzig, * Judge.

Opinion Text

Opinion By:

Miller, Judge.

This is an appeal from the decision of the Patent and Trademark Office ("PTO") Board of Appeals ("board") sustaining the rejection under 35 USC 103 of claims 9-13 and 15-16. We reverse.

Background

The Invention

Appellant's application ¹ discloses a process for the substantially *quantitative* production of hydroquinone. Claims 9-10 and 15 ² are directed to the reaction of *p*-isopropenylphenol and hydrogen peroxide in an inert solvent such as glacial acetic acid and in the presence of a catalytic amount of a strong mineral acid to produce hydroquinone and acetone (see reaction step (3) below). Claims 11-13 and 16 ³ are directed to

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the semicontinuous production of hydroquinone through the production of *p*-isopropenylphenol from phenol and acetone, according to the following reaction sequence:

Graphic material consisting of a chemical formula or diagram set at this point is not available. See text in hard copy or call BNA PLUS at 1-800-452-7773 or 202-452-4323.

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The Rejections and Appellant's Response

The examiner rejected claims 9-10 and 15 as unpatentable over a patent to Robert H. Saunders ("Saunders"), ⁴ which discloses a process for the preparation of various phenols from a,

b 1-unsaturated alkylsubstituted aryl ⁵ compounds. Hydroquinone is a phenol. ⁶ His reaction occurs in a liquid solvent, such as acetic acid, with a strong acid catalyst and, preferably, with "low cost" hydrogen peroxide as the oxidizing agent. In example 2, *a*-methylstyrene, which the examiner said is the closest in structure to the starting material of appellant's claimed invention (*p*-isopropenylphenol), is reacted with *t*-butylhydroperoxide to produce phenol in about an 80% yield:

Graphic material consisting of a chemical formula or diagram set at this point is not available. See text in hard copy or call BNA PLUS at 1-800-452-7773 or 202-452-4323.

Although the production of hydroquinone as such is mentioned by Saunders, the only suggested process begins with *a, a, a, a*

Tabular, graphic, or textual material set at this point is not available. Please consult hard copy or call BNA PLUS at 1-800-452-7773 or 202-452-4323.

t-tetramethyl-*p*-xylylene dialcohol as the starting material. ⁷

The examiner's rejection of claims 11-13 and 16 was based on Saunders in view of a patent to John L. Jones ("Jones"). ⁸ Jones was cited for the disclosure of a method for the preparation of phenols having unsaturated substituents (such as *p*-isopropenylphenol -- the starting material of claims 9-10 and 15) via a two-step reaction. In the first step, a phenol is condensed in the presence of an acid catalyst with a ketone to produce an intermediate "condensation product," which is then treated with a strong alkali metal base to neutralize the acidic catalyst. The mixture undergoes pyrolysis producing the desired phenolic compound (with an unsubstituted side chain), phenol, and a polymeric residue. On appeal, appellant has not argued that the first two steps of his reaction sequence, in which *p*-isopropenylphenol is produced, would not have been obvious in light of the PTO's citation to Jones.

In response to the rejections, appellant submitted several affidavits (including two by him) under 37 CFR 1.132. One by Sheng-Hong A. Dai described two tests. In the first test ("Dai I"), the procedure of ex

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ample 2 of the Saunders patent was followed "exactly" ⁹ (the starting material being *a*-methylstyrene), except that hydrogen peroxide (instead of *t*-butylhydroperoxide) was used as the oxidizing agent. A yield of only 15.8% phenol was reported; the major product was believed to be dimers of the *a*-methylstyrene. ¹⁰ In the second test ("Dai II"), the "exact" reaction conditions of example 2 of Saunders were followed except that *p*-isopropenylphenol (the starting material of appellant's process) was used. A yield of only 47% hydroquinone was obtained.

Appellant also directed the examiner's attention to a published German application, ¹¹ which discloses a process for the production of hydroquinone from 1,4-diisopropenylbenzene ¹² in the presence of glacial acetic acid as the solvent, either mineral acids or Friedel-Crafts type compounds as the catalyst, and hydrogen peroxide as the oxidizing agent. The pertinent examples

indicate yields of hydroquinone of about 10-35%.¹³

The Decision Below

The board found that the cited references "make out a strong case of prima facie obviousness" against appellant's claims, because (1) the starting material of the claimed process is encompassed in Saunders' generic starting materials; (2) the oxidizing agent in the claimed process (hydrogen peroxide) is stated by Saunders to be the preferred peroxide (due to its low cost); and (3) Saunders and Jones disclose all of the other reaction conditions. It further found that the disclosure of hydroquinone production by the Saunders process, "albeit from a different starting material than the one recited in appellant's claims, would lead one of ordinary skill to expect that the presence of the first formed hydroxyl group [on the benzene ring] would not interfere with the process of forming the second hydroxyl group." The board said the affidavit evidence was unpersuasive and concluded that the Carleton I and II affidavits failed to follow the prior art closely enough. It particularly criticized the concentration of the reactants and catalysts and the short reaction times.

On reconsideration, the board adhered to its original decision, saying that the affidavits merely indicate that it is possible to operate within the teachings of Saunders without obtaining a good yield. It also said that the Dai II result conflicts with the result in example 9 of appellant's specification, illustrating that there can be considerable variation in yield from "minor modification in reaction conditions."

Opinion

[1] We do not agree with the board's characterization of the showing made by the examiner as a "strong" prima facie case. In a 35 USC 103 case, the burden of proof is on the PTO to establish a prima facie case of obviousness. *In re Warner*, 54 CCPA 1628, 379 F.2d 1011, 154 USPQ 173 (1967), cert. denied, 389 U.S. 1057 (1968), and, once this has been accomplished, the burden of going forward with evidence to rebut that prima facie case is shifted to the applicant. *In re Murch*, 59 CCPA 1277, 464 F.2d 1051, 175 USPQ 89 (1972); *In re Hyson*, 59 CCPA 782, 453 F.2d 764, 172 USPQ 399 (1972). Whether a prima facie case is "strong" or "weak" is not material. If the applicant presents rebuttal evidence, the decision-maker must consider all of the evidence of record (both that supporting and that rebutting the prima facie case) in determining whether the subject matter as a whole would have been obvious.¹⁴ *In re Rinehart*, 531

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F.2d 1048, 189 USPQ 143 (CCPA 1976); *In re Lewis*, 58 CCPA 1270, 443 F.2d 389, 170 USPQ 84 (CCPA 1971). The correct procedure for considering rebuttal evidence was set forth by this court in *In re Rinehart*, *supra* at 1052, 189 USPQ at 147:

Though the burden of going forward to rebut the prima facie case remains with the applicant, the question of whether that burden has been successfully carried requires that the entire path to decision be retraced. An earlier decision should not, as it was here, be

considered as set in concrete, and applicant's rebuttal evidence then be evaluated only on its knockdown ability. Analytical fixation on an earlier decision can tend to provide that decision with an undeservedly broadened umbrella effect. Prima facie obviousness is a legal conclusion, not a fact. Facts established by rebuttal evidence must be evaluated along with the facts on which the earlier conclusion was reached, not against the conclusion itself. Though the tribunal must begin anew, a final finding of obviousness may of course be reached, but such finding will rest upon evaluation of all facts in evidence, uninfluenced by any earlier conclusion reached by an earlier board upon a different record.

The similarities between the processes of Saunders and those of appellant are evident, the only significant differences alleged by appellant being: (1) the starting material (Saunders uses A-methylstyrene, whereas appellant uses *p*-isopropenylphenol),¹⁵ and (2) the oxidizing agent (Saunders uses *t*-butylhydroperoxide, whereas appellant uses hydrogen peroxide).

Regarding (1), the board noted that the generic description of Saunders' starting materials ("unsaturated alkylsubstituted aryl compounds") encompasses appellant's starting material since the term "aryl" includes the phenyl radical. Assuming that a hydroxyl group on the benzene ring of Saunders' starting material (which is the only difference between the starting material used by appellant and that used by Saunders) would *not* interfere with the reaction process disclosed by Saunders, the board said that it would have been obvious to make hydroquinone (which differs from phenol only in the hydroxyl group at the *para*-position) from a starting material such as that used by Saunders, in which a hydroxyl group¹⁶ is attached at the *para*-position.

Regarding (2), the board noted that, although example 2 of Saunders uses *t*-butylhydroperoxide as the oxidizing agent, Saunders makes the general observation that hydrogen peroxide is the preferred oxidizing agent "due to its low cost." It concluded, therefore, that it would have been obvious to one skilled in the art to use hydrogen peroxide as the oxidizing agent in appellant's process. In view of the foregoing, we are persuaded that the board properly found that the prior art cited by the examiner established a prima facie case of obviousness.¹⁷ However, we are also persuaded that the board erred in failing to give proper weight to appellant's rebuttal affidavits, which directly attacked the premises on which the board based its determination of obviousness. Cf. *In re Lewis*, supra.

The only difference between Dai II and example 2 of Saunders is the presence in Dai II's starting material (the same as appellant's starting material) of a hydroxyl group on the benzene ring in the *para*-position.¹⁸ The board assumed that this difference would not interfere with the reaction process disclosed by Saunders. However, the yield of hydroquinone was dramatically less when appellant's starting material was reacted in Dai II according to Saunders' process (47%) than the yield of phenol obtained with Saunders' starting material (80%). Thus, the Dai II result contradicts both the board's assumption and its conclusion that "one of ordinary skill . . .

[would] expect that the presence of . . . [one] hydroxyl group would not interfere with the process of forming the second hydroxyl group." We are satisfied that one skilled in the art, viewing the Dai affidavit, would have concluded that the presence of such a hydroxyl group results in a significantly lower yield of hydroquinone than the amount that would be obtained with the Saunders' process, and that appellant's starting material could not be readily substituted in Saunders' process to achieve similar results.¹⁹

Appellant also relies upon the published German application²⁰ for its teaching that, under the process conditions of the appealed claims, both isopropenyl groups of *p*-diisopropenylbenzene are not readily converted to hydroxyl groups to yield hydroquinone.²¹ This teaching further supports appellant's position and contradicts the board's assumption that a first formed hydroxyl group would *not* interfere with the process of forming the second hydroxyl group; moreover, the low yields are in marked contrast to the substantially quantitative yields obtained by appellant.

Finally, appellant has shown by the Dai I affidavit that, despite Saunders' observation that hydrogen peroxide is the preferred oxidizing agent because of its low cost, it would not have been obvious to one of ordinary skill in the art to use hydrogen peroxide with the starting material of appellant's process. Dai I demonstrates that when hydrogen peroxide is used in Saunders' example 2, a yield of only 15.8% phenol is obtained in contrast to the 80% yield in Saunders' example 2. Accordingly, one skilled in the art would hardly have expected to obtain a substantially quantitative yield if hydrogen peroxide was the oxidizing agent of a different starting material from that used in Saunders' example 2.

[2]The board's response to appellant's rebuttal evidence was:

Appellant's showing that it is possible to operate within the reference disclosure without obtaining a good yield of the desired product is insufficient to refute the teachings of the reference, since they have presented no evidence of making experiments and adaptations which one of ordinary skill in this art would make as a matter of course if he did not immediately obtain the desired results.

Although in appropriate cases such a response might be persuasive,²² it overlooks that the two variations from the closest prior art in the tests reported by the affidavits (different starting materials and different oxidizing agents) were the very ones that the board relied on in finding a *prima facie* case of obviousness. As related above, the results of those tests would negate any expectation of one skilled in the art that these variations in the Saunders' process would result in a substantially quantitative production of hydroquinone. Although there is a vast amount of knowledge about general relationships in the chemical arts, chemistry is still largely empirical, and there is often great difficulty in predicting precisely how a given compound will behave. As the Second Circuit said in *Schering Corp. v. Gilbert*, 153 F.2d 428, 433, 68 USPQ 84, 89 (2d Cir. 1946):

while analogy is at times useful, organic [as well as inorganic] chemistry is essentially an experimental science and results are often uncertain, unpredictable and unexpected.

Accordingly, we hold that the affidavits, when considered with all of the evidence, are sufficient as a matter of law to rebut the prima facie case of obviousness.

The rejection of claims 9-13 and 15-16 is reversed.

Reversed.

Footnotes

Footnote 1. Serial No. 563,464, filed March 31, 1975, for "Process."

Footnote 2. Claim 9, which is illustrative of this set of claims, reads:

9. A process which comprises reacting p-isopropenylphenol with an at least equimolar amount of hydrogen peroxide in the presence of glacial acetic acid and a catalytic amount of a strong acid selected from the group consisting of sulfuric, phosphoric, p-toluenesulfonic, benzenesulfonic, methanesulfonic and ethanesulfonic acids, said reaction being carried out at a temperature not greater than 80°C whereby there is obtained hydroquinone and acetone.

Footnote 3. Claim 12, which is illustrative of this set of claims, reads:

12. A semicontinuous process for the conversion of phenol to hydroquinone which comprises condensing phenol and acetone in the presence of acid to obtain Bisphenol A, subjecting said Bisphenol A to alkaline hydrolysis to yield a mixture of phenol and p-isopropenylphenol subjecting said mixture of phenol and p-isopropenylphenol, without separation, to reaction with at least an equimolar amount, based on p-isopropenylphenol, of hydrogen peroxide in the presence of an inert solvent and a catalytic amount of a strong acid selected from the group consisting of sulfuric, phosphoric, p-toluenesulfonic, benzenesulfonic, methanesulfonic and ethanesulfonic acids said reaction being carried out at a temperature not greater than 80°C, to obtain a mixture of hydroquinone, phenol, and acetone, recovering the hydroquinone therefrom, and recovering the phenol and acetone generated as byproducts for re-use as starting materials in a subsequent cycle of the above steps.

Footnote 4. U.S. Patent No. 2,644,014, issued June 30, 1953, for "Phenol Production."

Footnote 5. The patent defines the term "aryl" to include the "phenyl" radical.

Footnote 6. A phenol is a benzene ring to which a hydroxyl group (-OH) has been attached. *Graphic material consisting of a chemical formula or diagram set at this point is not available. See text in hard copy or call BNA PLUS at 1-800-452-7773 or 202-452-4323.*

Other substituents (or groups) may be attached to the benzene ring at various points around the

ring. Hydroquinone is within the class of phenols because it has an additional hydroxyl group attached to the benzene ring:

Graphic material consisting of a chemical formula or diagram set at this point is not available. See text in hard copy or call BNA PLUS at 1-800-452-7773 or 202-452-4323.

Footnote 7. The process would be represented as follows:

Graphic material consisting of a chemical formula or diagram set at this point is not available. See text in hard copy or call BNA PLUS at 1-800-452-7773 or 202-452-4323.

Footnote 8. U.S. patent No. 2,497,503, issued February 14, 1950, for "Preparation of Substituted Phenols."

Footnote 9. A miscalculation in the original Dai affidavit was corrected before the board's opinion on reconsideration, thereby alleviating the board's concern in its original opinion that an excessive amount of sulfuric acid was used in Dai I.

Footnote 10. Although conditions were somewhat varied from those of the Saunders reference, the first affidavit of appellant Carleton ("Carleton I") reports the result of a similar experiment. No phenol was detected, and the major products were a dimer of -methylstyrene and 1,1,3-trimethyl-3-phenylindan.

Footnote 11. German patent application No. S33,841 (now patent No. 947,308), filed June 12, 1953, by Societe des Usines Chimiques Rhone-Poulenc and published on February 23, 1956.

Footnote 12. 1,4-diisopropenylbenzene has the following structure.

Graphic material consisting of a chemical formula or diagram set at this point is not available. See text in hard copy or call BNA PLUS at 1-800-452-7773 or 202-452-4323.

Footnote 13. The second affidavit of appellant ("Carleton II") reports an experiment similar to those of the German application in which 1,4-diisopropenylbenzene was also the starting material. A yield of about 28% of hydroquinone was obtained.

Footnote 14. The Solicitor's brief states that "the issue is whether the decision of the Board of Appeals was clearly erroneous." However, "obviousness is a legal conclusion based on factual evidence, *Graham v. John Deere Co.*, [383 U.S. 1, 148 USPQ 459 (1966)] . . . and not a factual determination." *In re Warner*, 54 CCPA 1628, 1634 n.6, 379 F.2d 1011, 1016 n.6, 154 USPQ 173, 177 n.6 (1967). Therefore, the proper issue before us is whether the board erred, as a matter of law, in holding that the claims were properly rejected under 35 USC 103. In deciding this issue the court will make "an independent determination as to the legal conclusions and inferences which should be drawn from . . . [the findings of fact]." See *United States v. Mississippi Valley Generating Co.*, 364 U.S. 520, 526 (1961).

Footnote 15. The structural difference is that appellant's starting material has a hydroxyl group on the benzene ring in the *para* -position to the isopropenyl group.

Graphic material consisting of a chemical formula or diagram set at this point is not available. See text in hard copy or call BNA PLUS at 1-800-452-7773 or 202-452-4323.

Footnote 16. Or, alternatively, a group, such as an isopropenyl group, which can be readily converted to a hydroxyl group by Saunders' process.

Footnote 17. As indicated earlier, appellant has not contended that Jones was improperly applied against claims 11-13 and 16.

Footnote 18. This difference of a hydroxyl group results in hydroquinone being the product in Dai II, rather than phenol, which is produced in Saunders' example 2.

Footnote 19. Contrary to the board's statement that the Dai II result conflicts with the result in example 9 of appellant's specification, the results are consistent. Under different reaction conditions (e.g., temperature of 105°C in Dai II vs. 30°C in example 9), example 9 reports only a 32% yield of hydroquinone. This substantiates the conclusion that a significantly lower yield is achieved when appellant's starting material is used in Saunders' process.

Footnote 20. Although the teachings of the published German application were argued before the board, it made no mention of the German application in its opinion.

Footnote 21. The low yields of about 10-35% are obtained even though hydrogen peroxide is used as the oxidizing agent. The result of Carleton II (28%) confirms the teaching of the German application.

Footnote 22. Cf. *In re Weber*, 56 CCPA 900, 405 F.2d 1403, 160 USPQ 549 (1969); *In re Michalek*, 34 CCPA 1124, 162 F.2d 229, 74 USPQ 107 (1947).

Footnote * The Honorable Robert L. Kunzig, Judge, United States Court of Claims, sitting by designation.

- End of Case -